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THEORY OF HIGH-TEMPERATURE MAGNETOSTRICTION

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White Oak, Silver Spring, MarylandTECHNICAL REPORT NO. 6
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In contrast to the rapid decrease predicted by conventional theory, the magnetostriction λ_{100} of iron has a large maximum just below the Curie temperature. We propose a mechanism based on the fact that an ellipticity in the quasi-particle spectrum permits a lowering of the free energy by distortion; an equivalent mechanism arises from the anisotropic magnon-phonon interaction near the zone boundary. This latter interaction is large at temperatures such that magnon renormalization (due to magnon-magnon interaction) lowers the magnon spectrum to degeneracy with phonons at the zone edge. The degeneracy temperature agrees well with the temperature of the λ_{100} maximum in iron. Adding silicone raises impurity states from the phonon spectrum and thence lowers the degeneracy temperature, but increases the range of temperature over which near-degeneracy occurs; this agrees with the observed shift and broadening of the λ_{100} peak. The degeneracy does not occur in nickel, nor does the λ_{100} peak. The mechanism also predicts a monotonically decreasing λ_{111} of the opposite sign to λ_{100} , as observed in iron.

*Supported in part by the Office of Naval Research.

MAY 21 1963

I. INTRODUCTION

The conventional magnetoelastic coupling theory of magnetostriction^{1,2} and the observations of Tatsumoto and Okamoto³ on iron are in puzzling disagreement. The magnetoelastic coupling theory predicts that the magnetostriction of ferromagnets should fall monotonically to zero with increasing temperature. In contrast the magnetostriction constant λ_{100} of iron, shown in Fig. 1a, increases with increasing temperature, exhibiting a minor maximum at about one-fifth of the Curie temperature and a major, large maximum just below the Curie temperature. Addition of small fractions of silicon shifts the major maximum to lower temperature and broadens it. The second magnetostriction constant λ_{111} , shown in Figure 1b, falls monotonically to zero. Finally, the peak in λ_{100} is absent in nickel.

In this paper we propose a mechanism which accounts qualitatively for the magnitude and location of the high-temperature maximum of λ_{100} in iron, for the shift and broadening of the maximum with the addition of silicon, for the absence of the effect in nickel, and provides criteria for the presence of the effect in other materials. Furthermore, the theory properly predicts the monotonic behavior of λ_{111} and the fact that λ_{111} and λ_{100} are of opposite sign.

The mechanism depends on the existence of an asymmetry in the excitation spectrum of the system, this asymmetry being determined by the axis of the magnetization. For clarity we first illustrate the effect by assuming a simple eccentricity of the spin-excitation (magnon) spectrum, such as results, for instance, from the dipolar interaction of the spins.⁴

Such an eccentricity is indicated schematically in Fig. 2, where the surfaces of constant frequency of the excitations are shown as elliptical.

The free energy of the crystal is the sum of individual contributions $F(\vec{k}, T)$ from the individual modes. The product of $F(\vec{k}, T)$ and of the density of modes in reciprocal space, $\frac{V}{(2\pi)^3}$, defines a scalar field in reciprocal space. This free energy density must be integrated over the Brillouin zone to compute the free energy of the crystal.

The surfaces of constant free energy density coincide with the surfaces of constant frequency of the modes, and an eccentricity of the latter implies a similar eccentricity of the former.

If the crystal is distorted, two effects must be considered. First, the boundaries of the Brillouin zone over which the free energy density is to be integrated, are shifted. And second, because of the change in interatomic distance the $\omega(\vec{k})$ dependence may be altered, in turn changing the free energy density field.

In the presence of an eccentricity the total free energy is decreased by a shift of the Brillouin zone boundary to exclude a region of reciprocal space corresponding to high free energy density in favor of another region of lower free energy density. This effect is indicated schematically in Fig. 2. Distortion of the Brillouin zone boundary corresponds to an inverse distortion of the crystal in real space, and thence to magnetostriction. The model calculation carried out in Section 2 indicates that the second effect (the dependence of $\omega(\vec{k})$ on strain) is somewhat smaller than the zone boundary effect, and that both effects are

of the same sign. Both effects are shown in Fig. 3, where the zone boundary effect is designated as a "surface" effect, and the shift in the frequencies throughout the zone is designated as the "volume" effect.

As indicated in Fig. 3, the magnetostriction resulting from a single eccentricity would be in qualitative agreement with the observations in iron if we were to choose the eccentricity opposite in sign and eight times the magnitude of that resulting from dipolar interactions. However, no plausible source of that type of eccentricity, with the required magnitude, is apparent to us. Consequently, as an alternative mechanism, we shall show in Section 3 that the magnon-phonon interaction near the Brillouin zone boundary produces an asymmetry in the excitation spectra which is equivalent in its effect to a simple eccentricity. The magnon-phonon interaction is itself sharply temperature dependent, becoming appreciable only when the magnon-magnon interaction renormalizes the magnon spectrum to lower the magnon frequencies so that they become degenerate with the phonons near the zone edge. This degeneracy temperature is just below the Curie temperature in iron. The resultant contribution to λ_{100} is therefore very small except in the neighborhood of the degeneracy temperature, leading to a strong maximum just below the Curie temperature, qualitatively similar to that observed in iron.

The systematics of the shift and change of shape of the λ_{100} maximum with addition of impurities, and the criteria for the presence of the maximum in other materials, then follow from the relative magnitudes of the unrenormalized magnon and phonon energies in these materials, and from the resultant possibility of magnon-phonon degeneracy near the zone edge.

II. SIMPLE ECCENTRICITY MODEL

Without specifying the source of the eccentricity at this stage, and simply to illustrate the effect in its most elementary form, we first assume that the magnon spectrum can be represented by

$$\omega(\vec{k}, \vec{\epsilon}) = \omega_0(\vec{k}, \vec{\epsilon}) + AYM \sin^2 \theta \quad (1)$$

where $\omega(\vec{k}, \vec{\epsilon})$ is the frequency of a mode of wave vector \vec{k} in an arbitrarily distorted crystal (indicated by the strain tensor $\vec{\epsilon}$), A is an undetermined dimensionless eccentricity constant, $YM = \frac{ge}{2mc}$ M is inserted to insure that the eccentricity vanish in the symmetric paramagnetic state, and θ is the angle between \vec{k} and \vec{M} . This simple eccentricity cannot be valid at very low \vec{k} , where it would lead to negative $\omega(\vec{k}, \vec{\epsilon})$, but the modes near the Brillouin zone boundary are of principal importance.

The dependence of $\omega_0(\vec{k}, \vec{\epsilon})$ on both \vec{k} and $\vec{\epsilon}$ is taken from the Green function theory of the Heisenberg model. Two versions of that theory have been given, by Tahir-Kheli and ter Haar,⁵ and by H. Callen.⁶ Tahir-Kheli and ter Haar find that the magnon energies are equal to simple spin wave energies multiplied by the fractional magnetization $m(T)$. The theory of Callen, while giving a somewhat more complicated renormalization which agrees with the Dyson result at low temperature, is not radically different from the magnetization renormalization at high temperatures. For simplicity, then, and because we are interested in the high temperature region, we simply adopt the magnetization renormalization. Then, assuming nearest neighbor interaction, the frequency (at a fixed point \vec{k} in reciprocal space) becomes

$$\hbar\omega_0(\vec{k}, \vec{\epsilon}) = 2JSm(T)(z - \sum_{\vec{\delta}} e^{i\vec{k} \cdot \vec{\delta}}) \quad (2)$$

$$= 2JSm(T)(z - \sum_{\vec{\delta}_0} e^{i\vec{k} \cdot \vec{\delta}_0}) + 2JSm(T) \sum_{\vec{\delta}_0} \vec{k} \cdot \vec{\epsilon} \cdot \vec{\delta}_0 \sin \vec{k} \cdot \vec{\delta}_0 \quad (3)$$

where $\vec{\delta}$ designates the vectors to the nearest neighbors (of which there are z), and $\vec{\delta}_0$ designates these vectors in the unstrained crystal.

The free energy of the system of magnons is

$$F = \sum_{\vec{k}} F_{\vec{k}} = \sum_{\vec{k}} \frac{1}{2} \hbar\omega(\vec{k}) + \beta^{-1} \sum_{\vec{k}} \ln(1 - e^{-\beta\hbar\omega(\vec{k})}) \quad (4)$$

and we are interested in the free energy difference $\Delta F = F(\vec{\epsilon}) - F(\vec{\epsilon} = 0)$ in the strained and unstrained crystal. Furthermore we are interested only in those terms in ΔF which contribute to the magnetostriction constants λ_{100} and λ_{111} , in that they involve both the direction of magnetization (or θ) and the strain $\vec{\epsilon}$; denoting these terms in ΔF by $\Delta F'$, we easily find

$$\Delta F' = \int_{\vec{\epsilon}} \left(\frac{\partial F}{\partial \omega} \right)_0 \delta\omega_{\theta} d\tau + \int_{B.Z.} \left(\frac{\partial^2 F}{\partial \omega^2} \right)_0 \delta\omega_{\theta} \delta\omega_{\epsilon} d\tau \quad (5)$$

$$= \hbar \int_{\vec{\epsilon}} \left[\frac{1}{2} + n(\omega, T) \right] \delta\omega_{\theta} d\tau - \beta \hbar \int_{B.Z.} n_{\vec{k}} (1 + n_{\vec{k}}) \delta\omega_{\theta} \delta\omega_{\epsilon} d\tau \quad (6)$$

where

$$\delta\omega_{\theta} = \Lambda \gamma M \sin^2 \theta \quad (7)$$

$$\delta\omega_{\epsilon} = 2J \langle S^z \rangle \sum_{\vec{\delta}_0} \vec{k} \cdot \vec{\epsilon} \cdot \vec{\delta}_0 \sin \vec{k} \cdot \vec{\delta}_0 \quad (8)$$

$$n(\omega, T) = [e^{\beta\hbar\omega(\vec{k})} - 1]^{-1} \quad (9)$$

and where the first integral is over the differential volume between the strained and unstrained zones, whereas the second integral is over the volume of the unstrained zone. The first term in Eq. (6) corresponds to the surface term shown in Fig. 2. This term has a zero-point value even when the Bose occupation numbers $n(\omega, T)$ are all zero. The second term in Eq. (6) describes the effect of the change of excitation frequencies, and hence of the free energy density, throughout the zone when the crystal is strained. A third term, corresponding to the change in density of states within the zone, has been omitted because, being fully symmetric, it makes no contribution to λ_{100} or λ_{111} . Calculation of the integrals is simplified by expanding strains, angular factors, and wave-vector components in Cubic Harmonics and extracting the fully symmetric products. We have carried out such an integration for a simple cubic lattice, performing the numerical integrations on the NOL 7090. A check on the calculations is provided by the exact cancellation of volume and surface terms of symmetry Γ_α , required by the invariance of $\mathbf{k} \cdot \mathbf{r}$ in the coordinate system moving with the modes. Then, minimizing the sum of $\Delta F'$ and the elastic strain energy, one finds the equilibrium strains, or magnetostriction. The resultant magnetostriction is shown in Fig. 3, in which we show the portion of λ_{100} ascribable to the surface and volume terms in Eq. (6). The temperature dependence of $m(T)$ has been taken from the empirical magnetization curves of Terry,⁷ and the elastic constants from the extrapolated data of Rayne and Chandrasekhar.⁸ These are plotted in Fig. 4. It will be noted that the decrease in $c_{11}(T) - c_{12}(T)$ accounts for most of

the broad λ_{100} peak on the simple eccentricity model. The volume contribution to λ_{100} is zero at $T = 0$ where all occupation numbers vanish, and it falls to zero again at T_c because of the renormalization of magnon energies (Eq. (8)). The surface contribution has a large zero point contribution, and it then increases with increasing temperature as the modes near the zone boundary become occupied. The single adjustable constant appearing in the results is the eccentricity constant A of Eq. (1); it has been chosen as $A = -16\pi$ in Fig. 3, so that the theoretical λ_{100} coincides with the measured value extrapolated to 0°K . For comparison, the value of A corresponding to the Holstein-Primakoff⁴ spectrum (i.e., the dipolar-induced eccentricity) is $+2\pi$.

The corresponding theoretical value of the shear magnetostriction λ_{111} is properly opposite to λ_{100} in sign, but its magnitude is only one third of the experimental value at 0°K , and it drops off monotonically but too slowly with increasing temperature.

III. MAGNON-PHONON INTERACTION

We now consider a specific mechanism which produces an asymmetry of the required magnitude in the excitation spectrum of a ferromagnet. Although this asymmetry is more complex than the simple eccentricity considered previously, its effect is qualitatively similar. The mechanism to be considered arises from magnon-phonon interaction.

The magnon and phonon spectra each approach the Brillouin zone boundary with zero normal slope, but the energies vary considerably over the face of the zone. The phonon frequencies have been measured by G. G. E. Low,⁹ who finds $h\omega_p = 0.48 \times 10^{-13}$ ergs at the [111] vertex; the phonon spectrum in this direction is shown in Fig. 5. The [111] vertex is the principal region of low magnon energy on the zone surface, and, as will develop subsequently, is therefore of particular interest to us. The unrenormalized (low temperature) magnon energies, given by Eq. (2), can be obtained sufficiently accurately from the Curie temperature and the Green function theory,^{5,6} whence $h\omega_m^0 = 1.50 \times 10^{-13}$ ergs at the [111] vertex in iron. The magnon spectrum is also shown in Fig. 5.

At low temperatures there is virtually no magnon-phonon interaction in iron except in the region of the crossing of the two spectra, deep within the zone. However, as the temperature increases, the spin waves energies renormalize. As indicated previously, at high temperature this renormalization is merely a multiplication of the spin wave frequencies by the fractional magnetization $m(T)$. Thus the magnon spectrum of iron becomes

degenerate with the phonon spectrum at the zone edge at that temperature T_p for which

$$m(T_p) = \omega_p / \omega_m^0 \quad (\text{if } \omega_p < \omega_m^0). \quad (10)$$

In actuality the phonon energies also renormalize, although probably to a lesser extent than the magnons. Accordingly, and in the absence of a quantitative theory, we shall neglect this effect.

Because of the magnetoelastic coupling, the phonon and magnon spectra mix, producing new modes displaced upward and downward by frequency shifts $\pm \delta\omega$. This spectral distortion then induces the zone boundary distortion somewhat as in the previous magnon spectrum ellipticity model.

The interaction term between magnons and phonons is dictated by symmetry considerations, and in the classical field representation can be written in the form¹⁰

$$\mathcal{H}_{mp} = \int [B^{ijmn} M^m M^n + D^{ijmnkl} \frac{\partial M^m}{\partial r^k} \frac{\partial M^n}{\partial r^l}] \epsilon^{ij} d\mathbf{r} \quad (11)$$

where the B's and D's are phenomenological coupling constants. The effect of the first term is dominant at low wave vector, where it gives rise to the conventional theory of magnetostriction and to the magnon-phonon crossing interaction, investigated most fully by Schlömann.¹¹ The second term in the integrand of Eq. (11) vanishes at zero wave vector, but strongly dominates near the zone edge. The order of magnitude of this term has been estimated by Kaganov and Tsukernik;¹⁰ because it arises from a modulation of the exchange integral they estimate it to be of the order of $J a^3 (ka)^2$. At the zone boundary, where $ka \approx \pi$, this would give a magnon-phonon interaction of approximately 2×10^{-13} ergs/ion, dominating the B term

by two orders of magnitude! This estimated magnon-phonon interaction would be as large, or larger, than the phonon energy itself, which would lead to an instability of the interacting spectra, and a phase transition. But the estimate of Kaganov and Tsukernik is a very rough one, and we shall assume only that the magnon-phonon matrix element for modes near the zone boundary is $\hbar\omega_i$, where ω_i is less than but of the same order as the phonon frequency ω_p .

The displaced frequencies of the interacting magnon and phonon modes are then determined by the secular equation

$$\begin{vmatrix} \omega_m - \omega & \omega_i \\ \omega_i & \omega_p - \omega \end{vmatrix} = 0 \quad (12)$$

The two roots are

$$\omega = \begin{cases} \omega_m \pm \delta\omega \\ \omega_p \pm \delta\omega \end{cases} \quad (13)$$

where the upper sign is to be taken if $\omega_m > \omega_p$ and the lower sign if $\omega_m < \omega_p$, and where

$$\delta\omega = -\frac{1}{2}|\omega_m - \omega_p| + \frac{1}{2}[(\omega_m - \omega_p)^2 + 4\omega_i^2]^{\frac{1}{2}} \quad (14)$$

Thus the displacement of the modes reaches a maximum value of ω_i when $\omega_m = \omega_p$, and it falls to $\frac{\omega_i}{2}$ when $\omega_m - \omega_p = \pm \frac{3}{2}\omega_i$.

The interaction matrix element ω_i is certainly strongly dependent on the angle between the wave vector of the modes and the magnetization, as has been shown at the cross-over region at low k by Schlömann.¹¹ It is obvious by symmetry that a spin wave propagating along the magnetization cannot couple to a longitudinal phonon. However, the coupling can be large for modes propagating

perpendicular to the magnetization direction. We thus summarize the above considerations by assuming that $\delta\omega(\theta)$ is a function of the angle θ between \vec{k} and \vec{M} :

$$\delta\omega(\theta) = g(\omega_m - \omega_p) \sin \theta \quad (15)$$

where g is of the order of ω_p , falling to half its value when $\omega_m - \omega_p \approx \pm \frac{1}{2}g(0)$.

Again the free energy of the crystal is decreased by a distortion of the Brillouin zone, excluding regions of small splitting in favor of regions of large splitting. As in Eq. (6) the surface contribution to the change in free energy is

$$\Delta F'' = \int_{\text{S}} [F_k(\omega_m + \delta\omega) + F_k(\omega_p + \delta\omega)] d\tau \quad (16)$$

At the degeneracy temperature this change in free energy becomes

$$\Delta F'' = -\beta \hbar^2 \int_{\text{S}} n_{\vec{k}} (1 + n_{\vec{k}}) g^2 \sin^2 \theta d\tau \quad (17)$$

Comparison of this equation with the surface term in Eq. (6) shows that the same value of magnetostriction at T_p will be obtained if g^2 and A are related by

$$A = \frac{-\beta \hbar \omega_m^0 g^2}{[1 - \exp(-\beta \hbar \omega_p)] \gamma M_0 \omega_p} \quad (18)$$

Consequently our previous result that a value of $A = -16\pi$ was required to fit the value of λ_{100} at T_p implies that this fit will be obtained with $\hbar g \approx 10^{-14}$ ergs. This required value actually is considerably less than the value estimated from the magnon-phonon matrix element.

We first estimate the position and shape of the peak in pure iron, and then discuss the effect of silicon alloying.

The peak temperature, from Eq. (10), and Fig. 5, is that temperature at which $m(T_p) = \frac{0.48}{1.5}$. From the experimental magnetization curve of Fig. 4 we find $T_p = 0.96 T_c \cong 1000^\circ\text{K}$. This figure, somewhat higher than the experimental value, is reduced by the observation that both the phonon and magnon frequencies vary somewhat over the zone surface. Hence magnon-phonon degeneracy is reached at different temperatures at different points on the surface. The observed maximum therefore should be broadened and depressed from our initial theoretical estimate.

In addition to this "geometrical" broadening, at least two other effects broaden the magnetostriction maximum. At high temperatures the lifetimes of the magnons and phonons become short, so that the excitations are themselves broadened considerably. Furthermore there is an intrinsic breadth of the maximum which arises from the fact that the magnons and phonons interact even at temperatures other than T_p .

We recall that $\delta\omega$ is a maximum at T_p , and that it falls to half its maximum at the temperatures $T_{\pm\frac{1}{2}}$ determined by

$$m(T_{\pm\frac{1}{2}}) = \frac{\omega_p + \frac{3}{2}\omega_i}{\omega_m} = m(T_p) \pm \frac{3}{2} \frac{\omega_i}{\omega_m}. \quad (18)$$

If we assume arbitrarily that $\omega_i \cong \omega_p$, then $m(T_{-\frac{1}{2}}) \cong 0.8$ and $m(T_{+\frac{1}{2}}) \cong 0$, whence $T_{-\frac{1}{2}}$ is roughly 800°K and of course $T_{\frac{1}{2}} \cong T_c = 1043^\circ\text{K}$. Like the peak temperature, these "half width" points are again too high, but the theory predicts correctly that the line will be broadened asymmetrically because of the shape of the magnetization curve, with the low temperature rise flatter than the sharp high temperature drop-off.

The magnon-phonon interaction mechanism also accounts naturally for the considerable effect of small additions of silicon. The position of the magneto-striction peak is determined by the phonon modes at the zone boundary, and these modes are radically perturbed by the addition of light impurities. In fact the tendency of such impurities is to split localized modes off the top of the phonon band; even if such localization is not accomplished, the modes of short wave-length ($\lambda \approx a$) are shifted upward in frequency. Consequently, the addition of silicon shifts the magnetostriction peak to lower temperatures, and broadens it by moving the peak to a region of smaller $\left| \frac{\partial m(T)}{\partial T} \right|$. These effects appear in Fig. 1.

If ω_m^0 is less than ω_p , rather than greater as in iron, no degeneracy temperature occurs. Magnon-phonon interaction will then give a contribution to the magnetostriction at zero temperature if $\omega_p - \omega_m^0 < \omega_i$, and this contribution will decrease with increasing temperature. This appears to be the case in nickel. Hard materials (high ω_p) with low Curie temperatures should not show a high-temperature magnetostriction maximum.

The optimum condition for the observation of the high-temperature magnetostriction maximum is that ω_m^0 be perhaps twice to five times as large as ω_p . For if ω_m^0 is enormously larger than ω_p the degeneracy temperature becomes practically identical to the Curie temperature, and the peak becomes very narrow. The situation in iron is apparently very close to the optimum conditions.

ACKNOWLEDGMENT

We are grateful for the assistance of Miss Ann Penn and to Neil McElroy in programming the calculations.

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FIGURE CAPTIONS

- Fig. 1a. Magnetostriction constant λ_{100} vs temperature, of iron and silicon iron, according to E. Tatsumoto and T. Okamoto. J. Phys. Soc. Japan 14, 1588 (1959).
- Fig. 1b. Magnetistraction constant λ_{111} vs temperature, of iron and silicon iron, according to Tatsumoto and Okamoto.
- Fig. 2. Zone boundary distortion effect.
An ellipticity in the magnon spectrum with respect to the magnetization direction creates an asymmetry in the free energy density field. The free energy is then lowered by extending the zone boundaries to include regions of low free energy and density and contracting the boundaries to exclude the high energy regions.
- Fig. 3. Magnetostriction constant λ_{100} as a function of temperature arising from the simple eccentricity model. The ellipticity coefficient has been adjusted to reproduce the experimental magnetostriction extrapolated to 0°K. The "surface" contribution, as indicated in Fig. 2, and the "volume" term, due to the change in energy of the magnons throughout the zone when the crystal is strained, are shown separately, as is their sum and the experimental curve of Fig. 1.
- Fig. 4. The experimental reduced magnetization of iron as a function of temperature, according to E. M. Terry, Phys. Rev. 30, 133 (1910). The elastic stiffness constants $c_{11}(T) - c_{12}(T)$ and $c_{44}(T)$ of iron have been measured by J. A. Rayne and B. S. Chandrasekhar, Phys. Rev. 122, 1714 (1961) from

FIGURE CAPTIONS - Cont.

4.2°K to 500°K. K. Nakamura, Sci. Rept. Tohoku Univ. 25, 364 (1955-57) measured the compliance constants of iron in the range $295^{\circ}\text{K} < T < 795^{\circ}\text{K}$. The stiffness constants employed by us are the data of Rayne and Chandrasekhar extrapolated parallel to the measurements of Nakamura (converted).

Fig. 5. Phonon energy is a function of propagation vector in the [111] direction in iron, according to G. G. E. Low, Proc. Phys. Soc. 79, 479 (1962). The unrenormalized (0°K) magnon energy vs propagation vector in the same direction in reciprocal space is also shown.











